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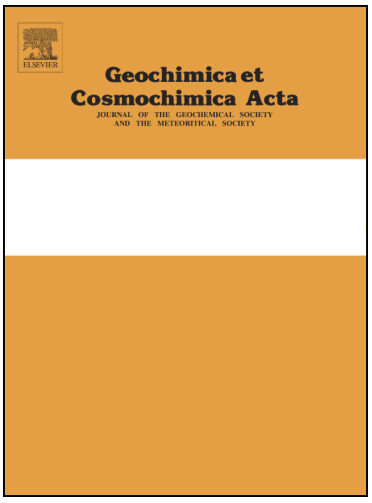

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Tellurium stable isotope fractionation in chondritic meteorites and some terrestrial samples

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ABSTRACT

New methodologies employing a ^{125}Te-^{128}Te double-spike were developed and applied to obtain high precision mass-dependent tellurium stable isotope data for chondritic meteorites and some terrestrial samples by multiple-collector inductively coupled plasma mass spectrometry. Analyses of standard solutions produce Te stable isotope data with a long-term reproducibility (2SD) of 0.064 ‰ for $\delta^{130/125}$Te. Carbonaceous and enstatite chondrites display a range in $\delta^{130/125}$Te of 0.9‰ (0.2‰ amu^{-1}) in their Te stable isotope signature, whereas ordinary chondrites present larger Te stable isotope fractionation, in particular for unequilibrated ordinary chondrites, with an overall variation of 6.3‰ for $\delta^{130/125}$Te (1.3‰ amu^{-1}). Tellurium stable isotope variations in ordinary chondrites display no correlation with Te contents or metamorphic grade. The large Te stable isotope fractionation in ordinary chondrites is likely caused by evaporation and condensation processes during metamorphism in the meteorite parent bodies, as has been suggested for other moderately and highly volatile elements displaying similar isotope fractionation. Alternatively, they might represent a nebular signature or could have been produced during chondrule formation.

Enstatite chondrites display slightly more negative $\delta^{130/125}$Te compared to carbonaceous chondrites and equilibrated ordinary chondrites. Small differences in the Te stable isotope composition are also present within carbonaceous chondrites and increase in the order CV-CO-CM-CI. These Te isotope variations within carbonaceous chondrites may be due to mixing of components that have distinct Te isotope signatures reflecting Te stable isotope fractionation in the early solar system or on the parent bodies and potentially small so-far unresolvable nucleosynthetic isotope anomalies of up to 0.27 ‰. The Te stable isotope data of carbonaceous and enstatite chondrites displays a general correlation with the oxidation state and hence might provide a record of the nebular formation environment.

The Te stable isotope fractionation of the carbonaceous chondrites CI and CM (and CO potentially) overlap within uncertainty with data for terrestrial Te standard solutions, sediments and ore samples. Assuming the silicate Earth displays similar Te isotope fractionation as the studied terrestrial samples, the data indicate that the late veneer might have been delivered by material similar to CI or CM (or possibly) CO carbonaceous chondrites in terms of Te isotope composition.

Nine terrestrial samples display resolvable Te stable isotope fractionation of 0.85 and 0.60‰ for $\delta^{130/125}$Te for sediment and USGS geochemical exploration reference samples, respectively. Tellurium isotopes therefore have the potential to become a new geochemical sedimentary proxy, as well as a proxy for ore-exploration.

1. INTRODUCTION

Tellurium (Te) is a chalcophile and siderophile element that is moderately volatile with a 50% condensation temperature (T_c) of 709 K similar to that of Zn (726 K) and Sn (704 K) (Lodders, 2003). Experimental data suggests that Te partitions strongly into the Earth's core (Rose-Weston et al., 2009), and therefore, it was suggested that a large portion of Te in the silicate Earth originates from the late veneer (Rose-Weston et al., 2009; Yi et al., 2000) consisting potentially of CI or CM carbonaceous chondrite-like material based on Se/Te ratios (Wang and Becker, 2013). However, the near-chondritic Se/Te ratio of the mantle may not be a primitive mantle signature, as has been suggested by (König et al., 2014). The isotope signature of Te in chondrites and in the terrestrial mantle is therefore expected to provide further constraints on the composition of the late veneer (Rouxel et al., 2002).

Non-traditional stable isotope fractionation data provide important information about the formation of the solar system, early solar system processes and the evolution of planetary bodies (e.g., Georg et al., 2007; Pogge von Strandmann et al., 2011; Poitrasson et al., 2004),

but Te stable isotope fractionation has scarcely been investigated. Terrestrial Te-rich ore minerals display significant fractionation of Te of 2‰ in $\delta^{130/125}$Te (Fornadel et al., 2017; Fornadel et al., 2014; Smithers and Krouse, 1968). Experimental data further demonstrates that abiotic and biotic redox processes can cause Te stable isotope variations (Baesman et al., 2007; Smithers and Krouse, 1968). Liquid-liquid extraction can induce both mass dependent (up to 1.9‰ in $\delta^{130/125}$Te) and mass independent Te isotope isotope fractionation (Moynier et al., 2008). In bulk meteorites, no mass-independent Te isotope variations are present indicating that the early solar system was well mixed and homogenous with respect to Te isotopes (Fehr et al., 2005). The study by Fehr et al. (2005) also derived low precision Te stable isotope fractionation data, and data for one sample, the unequilibrated ordinary chondrite Mezö-Madaras, indicated the possible presence of volatility induced fractionation of Te by evaporation and/or condensation processes during metamorphism. Ordinary chondrites display isotope fractionation for a series of moderately and highly volatile elements such as Ag, Rb, Zn and Cd that was most likely produced by metamophic processes on the meteorite parent body (Luck et al., 2005; Nebel et al., 2011; Schönbächler et al., 2008; Wombacher et al., 2008; Wombacher et al., 2003).

We have developed a new double spike procedure to obtain high precision Te stable isotope fractionation data of chondritic meteorites and some terrestial samples. These high precision Te isotope analyses will be used to determine if metamorphic processes cause volatility related fractionation of Te in ordinary chondrites. Additionally, we will investigate whether Te stable isotope fractionation has the potential to elucidate the origin of moderately and volatile elements on Earth and the nature of a potential late veneer addition by exploring whether compositional differences exist between different chondritic meteorite classes for Te isotopes.

2. SAMPLES

Bulk samples of 9 carbonaceous chondrites, comprising of the CI chondrite Orgueil, three CM chondrites (Cold Bokkeveld, Murray and Murchison), two CO chondrites (Ornans and Lance) and three CV3 chondrites (Allende, Grosnaja and Mokoja) were analysed to investigate their Te stable isotope fractionation. Additionally, two EH4 enstatite chondrites (Indarch and Abee) and six ordinary chondrites were measured: three LL chondrites (Bishunpur, LL3.1, Parnallee, LL3.6 and Tuxtuac, LL5), the L3.7 chondrite Mezö-Madaras and two H chondrites (Dhajala, H3.8 and Kernouve H6). For this study meteorite falls were exclusively studied to minimize the impact of terrestrial weathering.

Tellurium analyses of most terrestrial samples are more challenging compared to measurements of chondrites (Te contents of bulk chondrites: ~0.5-2.5 ppm, e.g. Fehr et al., 2005) due to their generally low Te concentrations. Most terrestrial crustal and mantle samples contain only a few ppb Te (e.g. Hein et al., 2003; Terashima, 2001). Eight terrestrial samples with known Te contents of 0.1 - 31 ppm Te (Table 1) were chosen in order to test our new analytical methodologies for a variety of sample matrixes and to start developing a framework for terrestrial Te stable isotope data. Additionally, the Te content and isotope composition of the USGS reference sample SDO-1 (Devonian Ohio shale) was determined. Six of the investigated terrestrial samples are sediments, including two manganese nodules, Nod-P-1 (Pacific ocean) and Nod-A-1 (Atlantic ocean), green river shale SGR-1 (Utah), marine mud Mag-1 (Gulf of Maine, Atlantic) and stream sediment JSD-2. A further three USGS geochemical exploration reference samples were also investigated: Jasperoid GXR-1 (Drum mountains, Utah), soil sample GXR-2 (Park City, Utah) and GXR-4 that is a mill heads sample of a porphyry copper ore from Utah (Allcott and Lakin, 1975).

3. ANALYTICAL METHODS

3.1. Sample preparation

Water from an 18.2 MΩ·cm -grade Millipore system and Teflon distilled mineral acids were used for all preparation. Sample powders of bulk chondrites (48 - 311 mg) and all terrestrial samples beside the two manganese nodules were digested in Savillex™ vials at 130°C using a series of dissolution steps. After each digestion step, the samples were dried down on a hotplate. First, the samples were dissolved in 5 ml of 29 M HF and 1.3 ml 14 M HNO_3 for 1 - 3 days. They were further digested with aqua regia, followed by subsequent digestion with 4.7 M HNO_3. Finally, the samples were dissolved in 6 M HCl. The two manganese nodules were dissolved using 6 M HCl. For double-spiked samples the ^{125}Te-^{128}Te double spike (see section 3.3) was either added to the sample powders before sample digestion or after completed digestion in the case where samples were split for analysis of both an un-spiked and spiked sample aliquot. Tellurium was then separated from sample matrixes using an anion-exchange separation procedure optimised after Fehr et al. (2004). Most matrix elements were separated from Te using the "HCl-method" detailed in Fehr et al. (2004) where samples are loaded in 2 M HCl onto 2 ml of AG1-X8 resin (200-400 mesh). The Te fractions were further purified using additional two separation steps using 1 ml and 0.2 ml of AG1-X8 resin. The purity of the final Te fractions was checked and tests using synthetic solutions were performed to ensure that any residual matrix elements present did not impact on the accuracy of the Te isotope data (Fehr et al., 2004, 2006, 2009). Yields of the total procedure are approximately 55 %. Total procedural blanks are ≤ 11 pg Te and are insignificant given that processed sample aliquots contain at least 26 ng Te.

3.2. Te isotope analyses

Tellurium isotope measurements were obtained with a Neptune multiple-collector inductively coupled plasma mass spectrometer (MC-ICPMS) housed at the Open University. Samples and standards were dissolved in 3% HNO_3 and introduced to the mass spectrometer using a PFA nebulizer with either a 20 or 50 μl min^{-1} flow-rate and an Aridus II desolvation nebulizer system. Normal sampler and X skimmer cones were used for analyses of sample and standard solutions containing 15- 30 ppb Te. Three sample analyses of manganese nodules were performed at 100 ppb Te using both standard sampler and skimmer cones. Ion beams were collected simultaneously on the Faraday cups at the following masses; 125 (^{125}Te), 126 (^{126}Te, ^{126}Xe), 128 (^{128}Te, ^{128}Xe), 129 (^{129}Xe), 130 (^{130}Te, ^{130}Xe, ^{130}Ba), and 135 (^{135}Ba). For both samples and standards we used the following measurement protocol. Before each analysis the system was washed with 3% HNO_3 for 5-10 minutes and on-peak backgrounds were then measured by collecting 40 ratios each with 4.2 s integration time, divided into 2 blocks of analyses. The sample or standard was then measured, with 100 Te isotope ratios collected each with 4.2 s integration time, separated into 5 blocks of analyses. Baselines of Faraday cups were collected with defocused ion beam for 30 s prior to each block of sample and standard measurements and at the start of each background analysis.

3.3 Tellurium double-spike procedure

To accurately characterise the stable isotope composition of Te, we developed a ^{125}Te-^{128}Te double spike methodology. The double spike composition was chosen to minimise analytical uncertainties based on error propagation in the double spike calculations, as well as to minimise interference corrections. Rudge et al. (2009) list a number of Te double spike combinations that are estimated to provide smaller uncertainties (by up to 35 % based on counting statistics) compared to utilising a ^{125}Te-^{128}Te double spike. However, many of these double-spike combinations use the less abundant Te isotopes that suffer from interferences

from Sn (^{120}Te, ^{122}Te, ^{124}Te) and Sb (^{123}Te), whereas the more abundant Te isotopes have interferences from the rare gas Xe (^{126}Te, ^{128}Te, ^{130}Te) and Ba (^{130}Te), with ^{125}Te being the only interference free Te isotope. Therefore, we chose to utilize the more abundant Te isotopes, ^{125}Te to ^{130}Te, which allows for a static data collection and avoids the smaller Te isotopes that suffer from Sn interferences and therefore minimises analytical uncertainties. Additionally, the use of a ^{125}Te-^{128}Te double spike with a ^{125}Te/^{128}Te ratio of 1.13 provides a small error magnification in the Te stable isotope determination over a large range of double spike – sample mixtures (Fig. 1).

While sample and standard analyses were background corrected using on-peak background measurements, small amounts of residual Xe and Ba required interference corrections for these elements and were performed using the measured abundance of ^{129}Xe and ^{135}Ba. Results are expressed as δ^{130}Te/^{125}Te = [(^{130}Te/^{125}Te $_{sample}$/ ^{130}Te/^{125}Te $_{Te\ Alfa\ Aesar\ metal}$)-1] × 1000 relative to a Te Alfa Aesar metal standard (Fehr et al., 2004). The double spike equations are solved using an iterative Newton Raphson-procedure assuming an exponential mass fractionation law to derive the fractionation factors (f) of the sample (nat) and double spike – sample mixture (mix), as well as the proportion (X) of double spike (sp) in the mixture (e.g. Albarède and Beard, 2004; Bonnand et al., 2011):

$$F^i\left(X_{sp}^{ref}, f_{nat}, f_{mix}\right) = X_{sp}^{ref} r_{sp}^i + \left(1 - X_{sp}^{ref}\right) r_{nat}^i \left(\frac{M^i}{M^{ref}}\right)^{f_{nat}} - r_{mix}^i \left(\frac{M^i}{M^{ref}}\right)^{f_{mix}} = 0$$

where r are the isotope ratios of the double spike, sample and mixture and M are the true masses of the isotopes (i; ^{125}Te, ^{126}Te, ^{128}Te) and the reference isotope ^{130}Te (ref).

The long-term reproducibility of the Te Alfa Aesar metal standard (154 analyses over 10 analytical sessions) is 0.064‰ for $\delta^{130/125}$Te, whereas internal errors (2 SE) of sample analyses vary from 0.017 to 0.053‰. Internal errors were related to measured signal intensities that varied with analyte concentrations and daily sensitivity. Errors for samples quoted in the manuscript and tables are 2 standard deviations (2 SD) of repeat analyses,

whereas for individual sample analyses the daily reproducibility of the Te standard is quoted as the uncertainty.

The Te concentration of the ^{125}Te-^{130}Te double spike was determined by inverse-isotope dilution relative to a gravimetrically prepared dilute solution of SRM 3156. Therefore, the Te concentrations in the samples can be determined by isotope dilution as a by-product of the Te isotope analyses.

3.4 Accuracy and precision of Te isotope and concentration data

The daily reproducibility of standard solutions ranged from 0.04 to 0.12‰ (2 SD) for $\delta^{130/125}$Te, whereas sample analyses that were typically repeated over several analytical sessions display slightly more variable results with a reproducibility of 0.02 – 0.2‰ (Tables 1 and 2). The number of repeat analyses is small though, where only 4 sample aliquots were measured ≥ 4 times. The reproducibily of $\delta^{130/125}$Te for different sample dissolutions varies from 0.003 to 0.58‰ and hence indicates to the presence of sample heterogeneities, in particular for Orgueil (2 SD of 0.42‰) and Mezö-Madaras (2 SD of 0.58‰), whereas other samples display reproducibilities of different dissolutions at ≤ 0.26‰. For all samples beside Orgueil (4 aliquots) and Allende (3 aliquots), only one or two separate sample aliquots were dissolved though. Analyses of Allende display a reproducibility of 0.17‰ for $\delta^{130/125}$Te.

Chondrite sample aliquots that were spiked before sample digestion display in general slightly lower Te contents by ~10% on average compared to sample aliquots that were spiked before sample digestion (Table 2, footnote e). This may potentially be indicative of Te loss during sample digestion. However, uncertainties of the Te concentration data are estimated to be up to several % reflecting relatively large uncertainties associated with weighing of small aliquots of the Te double spike. Furthermore, Te concentration data from the literature generally agree well with the data obtained in this study (Table 1, 2). Sample aliquots that

were spiked after sample digestion display only small differences in general and no systematic difference in the obtained $\delta^{130/125}$Te data compared to sample aliquots that were spiked before sample digestion (Table 2). Therefore, the data indicates that both spiking procedures provide accurate Te stable isotope data.

The accuracy of the employed methods was tested using three aliquots of the Te Alfa Aesar metal standard. Aliquots of 50 – 300 ng of Te standard were spiked with the ^{125}Te-^{128}Te double spike and processed using the anion-exchange separation procedure noted in section 3.1. Two column processed aliquots of the Te Alfa Aesar metal standard display small positive deviations from the unprocessed standard ($\delta^{130/125}$Te = 0) with up to 0.11 ± 0.08 ‰ for $\delta^{130/125}$Te (Table 1). On average, column processed aliquots of the Te Alfa Aesar metal standard display a $\delta^{130/125}$Te of 0.07 ± 0.10 ‰ that is identical to the unprocessed standard within uncertainty.

In bulk meteorite samples, no resolvable nucleosynthetic or radiogenic Te isotope anomalies are present (Fehr et al., 2005). Therefore, only a spiked sample analysis is in principle necessary to determine the Te mass dependent fractionation of samples. However, it is possible that small, so-far unresolvable, nucleosynthetic Te isotope variations exist. The effect of such potential anomalies on the obtained $\delta^{130/125}$Te results is investigated by varying the natural composition of the Te standard that is used in the deconvolution of the double spike equations based on the measured Te isotope variations for bulk chondrites of Fehr et al. (2005). These so far unresolvable potential nucleosynthetic Te isotope variations could produce a maximum variation of 0.27 ‰ in the measured $\delta^{130/125}$Te. Further analyses were performed in this study on a few selected un-spiked chondrite sample aliquots to verify the inexistence of nucleosynthetic and radiogenic Te isotope anomalies. These new data also indicate that the impact of nucleosynthetic anomalies onto the $\delta^{130/125}$Te results is ≤ 0.2 ‰.

The accuracy of terrestrial sample analyses was tested by analyses of un-spiked terrestrial sample aliquots, which have identical Te isotope compositions as standard solutions within analytical uncertainty.

4. RESULTS

4.1. Tellurium concentration data

Tellurium concentration data were obtained by isotope-dilution as part of the Te stable isotope analyses. For all samples beside SDO-1, literature Te concentration data are available and in general agree well with the data obtained in this study (Table 1, 2). The analysis of USGS reference sample SDO-1 gives a Te content of 0.12 ppm (Table 1) and hence SDO-1 has a similar Te content as the green river shale SGR-1 (0.2 ppm) and the marine mud rock Mag-1 (0.05-0.07 ppm).

4.2. Tellurium stable isotope results of terrestrial samples

The nine terrestrial samples analysed in this study show a range of 0.89‰ for $\delta^{130/125}$Te (0.18‰ amu^{-1}, Table 1, Figs. 2 and 3b) with values ranging from -0.15 ± 0.07 (GXR-2) to 0.74 ± 0.05 (Nod-A-1). The sediment and USGS geochemical exploration samples studied here cover a similar range in their Te stable isotope composition, beside Nod-A-1 that displays a slightly more positive $\delta^{130/125}$Te of 0.74 ± 0.05 ‰.

4.3. Tellurium stable isotope results of chondrites

The Te stable isotope composition of the investigated chondrites displays an overall variation of 6.3‰ for $\delta^{130/125}$Te (1.3‰ amu^{-1}, Table 2, Fig. 1), whereas the unequilibrated

ordinary chondrites Mezö-Madaras (L3.7) and Dhajala (H3.8) display the most extreme Te stable isotope signatures with $\delta^{130/125}$Te = -4.12 ± 0.58 and 2.15 ± 0.08, respectively. The Te isotope results obtained in this study for Mezö-Madaras using the double-spike methodology overlaps within uncertainties of previous results ($\delta^{130/125}$Te = -4.72 ± 2.25) obtained using a standard-sample bracketing technique (Fehr et al., 2005). There is no correlation of Te isotope fractionation with Te contents or petrographic types of ordinary chondrites (Fig. 4a). Carbonaceous chondrites show a smaller variation in the Te stable isotope composition at 0.78‰ for $\delta^{130/125}$Te (0.14‰ amu^{-1}), with Orgueil (CI) having the most positive $\delta^{130/125}$Te signature of 0.36 ± 0.17 for one of the investigated sample splits and an average of 0.04 ± 0.42 ‰ for 4 individually digested sample aliquotes. By contrast, Allende (CV3) and Lance (CO3.5) show the most negative $\delta^{130/125}$Te values obtained for carbonaceous chondrites with -0.30 ± 0.17 ‰ and -0.35 ± 0.20 ‰ on average, with one sample aliquot of Lance having the most negative $\delta^{130/125}$Te of -0.42 ± 0.05 ‰ (Table 2). The enstatite chondrites display a slightly more negative Te isotope signature compared to most carbonaceous chondrites with $\delta^{130/125}$Te = -0.56 ±0.07 for Abee (EH4) and $\delta^{130/125}$Te = -0.35 ± 0.08 for Indarch (EH4).

In summary, chondritic meteorites display a much wider range of stable Te isotope compositions (6.3‰ for $\delta^{130/125}$Te) than the small number of terrestrial samples (0.9‰ for $\delta^{130/125}$Te) studied here. Additionally, both enstatite chondrites and the CV and CO carbonaceous chondrites have more negative $\delta^{130/125}$Te compared to the terrestrial samples.

5. DISCUSSION

5.1. Tellurium stable isotope fractionation of terrestrial samples

Studies investigating Te isotope fractionation using high-precision isotope data of terrestrial samples are so far limited to Te ore minerals, mainly native Te, tellurides and tellurites, where Te isotope variations of 2‰ were observed for $\delta^{130/125}$Te (Fornadel et al., 2017; Fornadel et al., 2014). Furthermore, it was suggested that Te isotope fractionation of up to 4‰ in $\delta^{130/125}$Te can be generated between different Te ore minerals based on theoretical fractionation models (Fornadel et al., 2017; Smithers and Krouse, 1968). The three geochemical exploration reference samples analysed in this study also display resolvable Te isotope variations of 0.6‰ in $\delta^{130/125}$Te (Table 1, Fig. 2). Samples GXR-2 and GXR-4 contain sulfide minerals (Allcott and Lakin, 1975) that are likely the major host phases for Te, whereas sample GXR-1 is a jasperoid.

Tellurium distributions have not been studied in detail in sediments, although Schirmer et al. (2014) suggested the Te/Se ratio as a possible paleo-redox proxy. Significant Te isotope fractionation is evident in the terrestrial sedimentary reference samples with a range of 0.85‰ in $\delta^{130/125}$Te (Table 1, Fig. 2), highlighting the potential for Te isotopes as a sedimentary geochemical proxy. For many stable isotope systems, redox processes cause some of the largest isotope fractionation (e.g., Beard and Johnson, 2004) and hence might be the cause for some of the observed Te stable isotope variations, consistent with experimentally produced Te isotope fractionation (Baesman et al., 2007; Smithers and Krouse, 1968). The presented data in particular provide the first evidence for significant Te isotope variations in the marine environment based on results for two shales, a mud-rock and two ferromanganese nodules. Ferromanganese crusts and nodules have been used as palaeoceanographic archives that record changes in the composition of seawater through time (e.g., Frank, 2002) and are exceptionally enriched in Te (Hein et al., 2003). As tetravalent and hexavalent Te species can both exist in seawater (Lee and Edmond, 1985), Te isotopes can therefore potentially be used as an oceanographic redox proxy. Estimates of the residence time of Te in the ocean have large uncertainties since information on Te contents of distinct

Te sources are sparse. However, it was suggested that Te is likely to have a very short residence time in the ocean of around 100 years (Hein et al., 2003) and hence it is conceivable that ferromanganese crusts and nodules reflect the local seawater composition of Te. Therefore, our new Te isotope data for ferromanganese nodules may suggest the presence of variations in the Te isotope composition of inputs to the ocean. On the other hand, isotope fractionation upon incorporation of Te into ferrmonanganese crusts and nodules may occur, as has been shown for Mo and Tl (Barling et al., 2001; Rehkämper et al., 2002). Further studies will be necessary to investigate the cause for the observed Te isotope variations in sediments.

5.2. Tellurium stable isotope variations in chondrites

Tellurium as a moderately volatile element is depleted in different carbonaceous chondrite groups compared to CI chondites (e.g. Larimer and Anders, 1967), whereas ordinary chondrites are further depleted in moderately volatile elements compared to carbonaceous chondrites (e.g. Kallemeyn et al., 1989). The origin of these depletions is generally considered to reflect nebular (e.g. Larimer and Anders, 1967; Palme et al., 1988) and potentially parent-body processes (e.g. Wombacher et al., 2008) and the new Te stable isotope fractionation data are used to inform the different proposed formation scenarios.

Shock heating has been suggested to potentially contribute toward the loss of moderately volatile elements (Friedrich et al., 2004) and isotope fractionation in ordinary chondrites. The ordinary and carbonaceous chondrites investigated here are generally only mildly shocked. Our data indicate that shock-grades of up to 3 induce only minimal Te isotope fractionation, since Grosnaja (S3) has a $\delta^{130/125}$Te within analytical uncertainty of two CV3 carbonaceous chondrites that show no evidence for shock (S1). Further investigations will be necessary to investigate the effect of higher shock-grades on the Te stable isotope fractionation. Abee is the only sample that might have experienced slightly higher shock at

stage S2-4. Since vapor phases are generally enriched in the light isotopes compared to solid phases (e.g., Richter et al., 2002), as has also been suggested by theoretical calculations for Te (Fornadel et al., 2017; Smithers and Krouse, 1968), loss of Te due to strong shock heating could potentially have induced a positive shift of $\delta^{130/125}$Te in the shocked meteorite. All beside one of the investigated chondrites (Mezö-Madaras, L3.7) display more positive $\delta^{130/125}$Te values compared to Abee and hence the Te stable isotope composition of Abee is likely also unaffected by shock.

5.2.1. Tellurium stable isotope variations of carbonaceous and enstatite chondrites

5.2.1.1. Tellurium stable isotope fractionation of Orgueil

Individually dissolved sample aliquots of ~50 mg of Orgueil were investigated in this study, whereas Orgueil displays heterogeneities in trace element abundances for sample sizes of <200 mg (Barrat et al., 2012). However, concentrations of Te varied only slightly from 2.18 to 2.31 ppm for the four analysed sample aliquots of Orgueil (Table 2) and display no correlation with the $\delta^{130/125}$Te data. Tellurium is a chalcophile element that also displays siderophile behavior and hence there are indications from sequential leach experiments and analyses of different components of carbonaceous chondrites (Fehr et al., 2006; Kadlag and Becker, 2016b) that the major host phases of Te in carbonaceous chondrites are sulfides and metal. Heating experiments also suggest that Te is present in two phases within carbonaceous chondrites (Matza and Lipschutz, 1977). Since virtually no metal is present in Orgueil, Te is likely associated mainly with the sulfide mineral pyrrhotite (Bullock et al., 2005), although small abundances of cubanite ($CuFe_2S_3$; Kerridge et al., 1979), another potential sulfide host phase of Te, have also been reported. In contrast to other CI chondrites, Orgueil only contains pyrrhotite, but not pentlandite. Orgueil experienced a high degree of secondary alteration during which primary pendlandite is thought to be completely replaced by pyrrhotite (Bullock

et al., 2005). There are also indications that sulfate may be another significant Te-bearing phase within Orgueil based on Te contents of hot water leaches (Reed and Allen, 1966; Fehr et al., 2006). The majority of sulfate in Orgueil is considered to have formed during terrestrial sample storage (Gounelle and Zolensky, 2001) from primary sulfate and/or oxidation of sulfides (Gounelle and Zolensky, 2001; Kerridge et al., 1979). Furthermore, sulfur-bearing organic phases are present in carbonaceous chondrites (Gao and Thiemens, 1993a). Therefore, part of the chalcophile element Te may also be associated with organics in Orgueil. The differences in the Te stable isotope composition of different sample fractions of Orgueil (Table 2, Fig. 3) suggest the presence of phases with distinct $\delta^{130/125}$Te that are heterogeneously distributed in Orgueil. Pyrrhotite, cubanite, pentlandite, sulphate and organic phases as potential Te host phases in Orgueil may display distinct Te stable isotope signatures and hence the observed Te stable isotope variations of different sample aliquots of Orgueil could be explained by heterogenously distributed Te-bearing minerals. In contrast, different sample splits of Allende display smaller variations in their Te isotope composition, suggesting that aqueous alteration may be potentially responsible for the heterogeneity of the Te stable isotope compositions in Orgueil. Alternatively, Te might get redistributed by metamorphism, which could have affected in particular CO3 and CV3 carbonaceous chondrites and hence Allende. However, further investigations will be necessary to determine whether sample heterogeneity in terms of Te stable isotope variations increase with degree of aqueous alteration and lower metamporphic grade in carbonaceous chondrites, or whether Te stable isotope composition and Te contents of different Te host phases of Orgueil and other chondrite samples affect the observed Te stable isotope variations.

5.2.1.2. Depletion of moderately volatile elements

The concentrations of moderately and highly volatile elements decreases in carbonaceous chondrites from CI to CM, CO and to CV chondrites (e.g., Palme et al., 1988).

This depletion is thought to reflect incomplete condensation in hot areas of the nebula, where more volatile elements condensed only partly due to their lower condensation temperature compared to more refractory elements (e.g., Palme et al., 1988). Chondrites are a mixture of chondrules, which experienced melting and are generally depleted in volatile elements and matrix that consists of material that experienced less processing in the nebula and is enriched in volatile elements (e.g. Zanda, 2004; Jones et al., 2005). Tellurium is also depleted in the chondrules of Allende and Murchison compared to matrix fractions (Kadlag and Becker, 2016b). The abundance of chondrules increases with higher depletion of Te and other moderately volatile elements in carbonaceous chondrites (Kadlag and Becker, 2016b; Krot et al., 2003). Hence, chondrule formation may be responsible for the depletion of moderately volatile elements in different carbonaceous chondrites groups compared to CI chondrites (e.g., Alexander et al., 2001). The mass dependent Te isotope composition displays a general correlation with the Te content in carbonaceous chondrites (Fig. 4b), with one aliquot of the CI chondrite Orgueil displaying the most positive $\delta^{130/125}$Te signature, whereas three additional Orgueil sample splits have a Te stable isotope composition close to 0. Isotopically light Te is expected to be enriched in volatile components (Fornadel et al., 2017; Smithers and Krouse, 1968). Therefore, Te depletions due to volatile loss or incomplete condensation of Te would generally be expected to produce positive $\delta^{130/125}$Te values in different carbonaceous chondrite groups compared to CI. Hence, the observed Te stable isotope fractionation is unlikely caused by the process that is responsible for the depletion of Te. Similar correlations of moderately volatile element depletion with stable isotope variations of Zn and Cu were reported and associated with compositional differences within carbonaceous chondrites due to mixtures of two meteorite components (Luck et al., 2005), as will be further discussed in the next section.

5.2.1.3. Oxidation state and variations in the solar nebula

Sulfide phases are expected to be important carriers of Te in chondrites (Allen and Mason, 1973; Fehr et al., 2006; Kadlag and Becker, 2016a). However, empirical and theoretical data for Te isotope fractionation in sulfide phases are not available. In magmatic terrestrial systems, Te is highly incompatible in monosulfide solutions and may exist as Te (0) in sulfides (Helmy et al., 2010). Tellurium is expected to be chalcophile in Orgueil (as no metal is present) and have both siderophile and chalcophile tendencies in carbonaceous chondrites (Fehr et al., 2006; Kadlag and Becker, 2016b), whereas Te is mainly siderophile in enstatite chondrites (Kadlag and Becker, 2015). Hence, Te behaves in an inceasingly siderophile manner with decreasing oxidation states of chondrites, whereas enstatite chondrites are more reduced compared to the carbonaceous chondrites. Hence, the overall trends of mass dependent Te isotope composition displays a general trend with the oxidation state of carbonaceous and enstatite chondrites. Ordinary chondrites that are intermediate in oxidation state between carbonaceous and enstatite chondrites display a large spread in Te stable isotope variations, as will be discussed in the following section (5.2.2). On average, ordinary chondrites have a Te stable isotope composition that is intermediate between carbonaceous and enstatite chondrites with -0.4 ± 4.1 in $\delta^{130/125}$Te and also fit into the trend of Te stable isotope composition versus oxidation state. Isotopically heavy Te is generally expected to be enriched in more oxidized mineral phases (Fornadel et al., 2017) and could therefore explain the observed difference between enstatite and carbonaceous chondrites. This could also explain the small differences between different groups of carbonaceous chondrites and the most positive $\delta^{130/125}$Te values for the most oxidized sample Orgueil. However, aqueous alteration and parent body metamorphism might also have affected the Te stable isotope signature of carbonaceous chondrites. Nevertheless, the observed differences in the Te stable isotope composition of different carbonaceous and enstatite chondrite groups might be a nebular signature that reflects different redox environments within the early solar system.

Different chondrite groups and different carbonaceous chondrite classes display distinct nucleosynthetic isotope anomalies for several refractory elements, such as Cr, Zr and Mo, providing evidence for mixtures of two or more components in the early solar system (e.g., Dauphas et al., 2002; Trinquier et al., 2007; Burkhardt et al., 2011; Akram et al., 2015). Different groups of chondrites display different oxidation states and are expected to have formed in different regions of the solar nebula (e.g. Brearley and Jones, 1998) and potentially at different heliocentric distances (e.g. Rubie et al., 2015), as is also reflected in, for example, their Ru mass-independent isotope signature (Fischer-Gödde et al., 2017). Compositional variations of refractory elements and volatile elements might be linked as is evident by correlations of Cr with O mass-independent isotope variations (Trinquier et al., 2007). Additionally, variations in the Cu and Zn isotope stable isotope fractionation also display a correlation with mass-independent O isotope variations in carbonaceous chondrites (Luck et al., 2005). Luck et al. (2005) suggested that these Cu and Zn isotope variations reflect mixtures of two meteorite components within carbonaceous chondrites. According to Luck et al. (2005), one of these components could be represented by refractory inclusions that would be depleted in moderately volatile elements and enriched in the light isotopes due to reaction with an isotopically light gas phase. Tellurium is similar to other moderately volatile elements and is generally depleted in refractory inclusions compared to bulk samples (Brennecka et al., 2016; Fehr et al., 2009). The available Te isotope fractionation data for refractory inclusions from Allende though have large uncertainties at >10‰ for $\delta^{130/125}$Te and display no resolvable fractionation (Fehr et al., 2009). Hence, further work will be necessary to determine the origin of the small Te stable isotope variations of different chondrites groups to identify meteorite components with distinct compositions. Such a model would also be valid if small so far unresolved nucleosynthetic Te isotope variations would contribute to the differences in $\delta^{130/125}$Te ($\leq$ 0.27 ‰; section 3.4). Sulfur and Se are both chalcophile and moderately volatile elements that are geochemically akin to Te. Importantly, S displays also small compositional

differences in its mass dependent isotope composition for different chondrite groups (Gao and Thiemens, 1993b), whereas Se displays no systematic variations with present analytical uncertainties of around 0.1 ‰/amu (Vollstaedt et al., 2016). While further investigations combining high precision Te isotope analyses of spiked and unspiked sample aliquots will be necessary to verify how much nucleosynthetic variation exists, our data indicate that there are only small variations in the Te mass dependent stable isotope composition of carbonaceous chondrites.

5.2.2. Tellurium stable isotope variations in ordinary chondrites

Unequilibrated ordinary chondrites are the least modified meteorites, which contrasts with other chondrite groups whose composition is modified by metamorphism and aqueous alteration. Unequilibrated ordinary chondrites display larger Te stable isotope fractionation compared to ordinary chondrites of higher metamorphic grades with both positive and negative fractionation compared to most chondrites (Table 2, Fig. 3, Fig. 4). These large variations are not only manifested in Mezö-Madaras (L3.7) and Dhajala (H3.8), which display the most extreme Te stable isotope results. Parnallee (LL3.6) and Bishunpur (LL3.1) also display a spread of 0.87‰ in $\delta^{130/125}$Te, whereas Tuxtuac (LL5) and Kernouve (H6) have $\delta^{130/125}$Te values that overlap with the Te isotope composition of CV chondrites. The unequilibrated and equilibrated ordinary chondrites analysed in this study display very similar depletions of Te with an average of 0.28 ± 0.12 and 0.31 ± 0.03 ppm (Table 2), respectively. Similar systematics in isotope fractionation are also observed for the highly volatile element Cd (Wombacher et al., 2008). In contrast, Zn displays more isotope variability in equilibrated ordinary chondrites (Luck et al., 2005). It is conceivable that primitive isotope signatures were retained in unequilibrated ordinary chondrites and were erased by homogenization and redistribution of Te during secondary processes such as metamorphism and aqueous alteration

in other chondrite groups. Tellurium is expected to condense into the metal phase and possibly into troilite during condensation from gas of solar composition (Lodders, 2003). Hence, metal is an important host phase for the chalcophile element Te in addition to sulfides, as is supported by geochemical analyses of different ordinary chondrite components (Allen and Mason, 1973; Kadlag and Becker, 2016a; Mason and Graham, 1970). Tellurium seems to display variable behaviour in different unequilibrated ordinary chondrites with primarily siderophile behaviour in three investigated samples including Parnallee (Kadlag and Becker, 2016a; Kadlag and Becker, 2017) and predominantly chalcophile behaviour in one sample. The majority of primary metal and sulfide phases are expected to have been modified during chondrule formation and metamorphism (e.g., Scott and Krot, 2014) and therefore the Te stable isotope variations of ordinary chondrites is unlikely to be an undisturbed primary nebular signature.

5.2.2.1. Chondrule formation

Tellurium and other moderately volatile elements are depleted in chondrules relative to matrix (Bland et al., 2005; Kadlag and Becker, 2016a) and hence chondrule formation may be an important process that depleted those elements in chondrites and in particular in ordinary chondrites with their high abundance of chondrules (e.g., Larimer and Anders, 1967). Chondrules display no resolvable isotope fractionation for other moderately volatile elements including K (Alexander et al., 2000). In contrast, chondrules of unequilibrated ordinary chondrites display isotope fractionation of 2 – 3 ‰/amu for the highly volatile element Cd (Wombacher et al., 2008). Matrix, chondrules and bulk samples however, display very similar amounts of Cd isotope fractionation, indicating that the isotope signature of chondrules and matrix was produced in the same environment. Therefore, the observed isotope fractionation for Te and other moderately and highly volatile elements in ordinary

chondrites is not likely to be produced by chondrule formation. Further studies investigating Te isotope fractionation in chondritic components are necessary to verify this conclusion.

5.2.2.2. Parent-body metamorphism

Metal and sulfides that are likely the main carrier phases of Te in ordinary chondrites (Kadlag and Becker, 2016a; Mason and Graham, 1970) are probably redistributed during parent-body metamorphism (Grossman and Brearley, 2005; Zanda et al., 1994). It has been suggested that the depletion of moderately and highly volatile elements in ordinary chondrites is caused by parent body processes, where elements may be mobilised during metamorphism (e.g., Schaefer and Fegley Jr., 2010). Tellurium is not systematically depleted in higher metamorphic grade ordinary chondrites though compared to ordinary chondrites of lower metamorphic grades (Kallemeyn et al., 1989; Wolf and Lipschutz, 1998), as is also supported from the data obtained in this study (Table 2, Fig. 4a). There are indications though that Te concentrations in ordinary chondrites are more variable compared to the moderately volatile elements Se and Zn (Schaefer and Fegley Jr., 2010) for example. Tellurium stable isotope fractionation data of ordinary chondrites display no trend with Te concentrations (Fig. 4a), whereas Te contents vary considerably in the investigated samples from 0.19 to 0.35 ppm (Table 2).

Heating experiments indicate that a large proportion of the Te budget of ordinary chondrites is lost at temperatures of 873 K and more (Ikramuddin et al., 1977), whereas there is also evidence for a Te phase that releases Te at lower temperatures (673 K; Lauretta et al., 2002). Furthermore, theoretical calculations predict that half of the Te budget gets mobilised at 572 K (at 10^{-4} bar) – 757 K (at 1 bar) during metamorphism (T_m: 50% metamorphic temperature) of ordinary chondrites (Schaefer and Fegley Jr., 2010), whereas the 50% condensation temperature of Te is 709 K (Lodders, 2003). Particularly important is the thermal stability of the Te host-phases as was indicated by the distribution of Zn and Se and

their isotope fractionation in ordinary chondrites (Vollstaedt et al., 2016). Hence, given that metamorphic temperatures in unequilibrated ordinary chondrites are estimated to range from around 473 K to up to 873-973 K (e.g., Huss et al., 2006), Te is expected to be mobilised during metamorphism on the ordinary chondrite parent bodies and some Te may also have been lost or redistributed even from unequilibrated ordinary chondrites. In contrast, in some unequilibrated ordinary chondrite components, Te displays correlations with the siderophile refractory element Ir and with Pd, which may suggest that Te was not affected by metamorphism (Kadlag and Becker, 2017). Hence, this may indicate that the Te stable isotope signatures in unequilibrated chondrites might be a primary nebula signature or produced by chondrule formation. However, sulphide and metal phases are often inter-grown in chondrites and consist of multiple primary and secondary phases (e.g. Brearley and Jones, 1998; Scott and Krot, 2014), which complicates the interpretation of element abundance patterns of chondrite components.

Highly volatile and some moderately volatile elements including Ag, Cd, Rb and Zn display isotope fractionation in ordinary chondrites thought to be produced by metamorphic parent-body processes (Luck et al., 2005; Nebel et al., 2011; Schönbächler et al., 2008; Wombacher et al., 2008; Wombacher et al., 2003). The present study confirms that stable isotope fractionation is also evident for Te in unequilibrated ordinary chondites, as was initially suggested by Fehr et al. (2005). Tellurium (T_m = 572 K; T_c = 709 K) displays slightly more isotope variations overall with 1.3‰ amu^{-1} compared to the moderately volatile elements Zn (T_m = 840 K; T_c = 726 K;), Ag (T_m = 875 K; T_c = 996 K) and Rb (T_m = 675 K; T_c = 800 K) that have slightly higher 50% metamorphic and condensation temperatures at 10^{-4} bar (Lodders, 2003; Schaefer and Fegley Jr., 2010) and display 0.5 – 0.7 ‰ amu^{-1} fractionation (Luck et al., 2005; Nebel et al., 2011; Schönbächler et al., 2008). Selenium has a condensation temperature of 697 K (Lodders, 2003) and a slightly higher 50% metamorphic temperature of 626 K compared to Te at 10^{-4} bar (Schaefer and Fegley Jr., 2010) but displays

no resolvable isotope fractionation (<0.2‰ amu^{-1}; Vollstaedt et al., 2016). At 1 bar, Te (T_m = 757) is estimated to be significantly more mobile during metamorphism compared to Se (T_m = 875 K; Schaefer and Fegley Jr., 2010). Furthermore, ordinary chondrites display non-significant isotope variability for the moderately volatile element S (Gao and Thiemens, 1993b), which is estimated to be less mobile compared to Te during metamorphism (Schaefer and Fegley Jr., 2010). In contrast, larger isotope variations are reported for the highly volatile element Cd (T_m = 547 K; T_c = 652 K; Lodders, 2003; Schaefer and Fegley Jr., 2010) at 5.8‰ amu^{-1} (Wombacher et al., 2008).

This non-systematic relationship of isotope fractionation versus depletion in concentration and metamorphic grade can be explained by multiple-stage evaporation and re-condensation of moderately to highly volatile elements in colder parts of the ordinary chondrite parent bodies (Schönbächler et al., 2008; Wombacher et al., 2008). In an onion-shell model, the colder outer part of the parent bodies are considered highly porous facilitating loss of volatile elements as well as re-condensation, whereas the hotter inner regions are regarded as less porous allowing for higher degrees of back-reactions of volatiles (Schönbächler et al., 2008; Wombacher et al., 2008). Importantly, such a model can explain both positive and negative isotope fractionation, as is evident for Te (Fig. 3a). Open-system metamorphism associated with higher porosity and lower internal pressure would require lower temperatures for Te mobilisation compared to closed-system metamorphism (Schaefer and Fegley Jr., 2010). Hence, it is likely that the stable isotope fractionation of Te and other moderately and volatile elements in ordinary chondrites is caused by evaporation and condensation processes during metamorphism on the meteorite parent body. However, further work will be necessary to verify this conclusion and exclude chondrule formation and nebular processes as potential origin for the observed Te stable isotope variations.

5.3. The composition of the Earth and the late veneer

The solar system is relatively homogeneous in terms of elemental and isotopic compositions, although small variations between different planets and meteorite parent bodies exist (Palme et al., 2014). These small compositional differences provide important information on the building blocks of the Earth, which in the past were considered to have a similar chemical and isotopic composition to chondritic meteorites. Enstatite chondrites provide a match to the Earth in terms of nucleosynthetic isotopic composition for a number of elements (e.g., Dauphas and Schauble, 2016), whereas in terms of bulk elemental abundances, the composition is closer to carbonaceous chondrites (Palme and O' Neill, 2014). The initial accreting material to the Earth may have been more reduced and later accretion may have involved more oxidized and volatile rich material from the outer solar system (e.g. Wood et al., 2006; Schönbächler et al., 2010). This oxidized, carbonaceous chondrite like material is also expected to have delivered a large part of the moderately volatile element budget to the Earth (Schönbächler et al., 2010). There are also indications, for example, from nucleosynthetic isotope variations of Mo and Ru in chondrites that chondrites cannot have been the main constituents of the Earth (Dauphas et al., 2002; Burkhardt et al., 2011; Fischer-Gödde and Kleine, 2017). Therefore, it is conceivable that the building blocks of the Earth might be missing from our meteorite collection because they were accreted during terrestrial planet formation (Dauphas and Schauble, 2016), although material with near-chondritic composition could still make up large parts of the Earth (e.g. Dauphas, 2017).

Core formation has depleted the silicate portion of the Earth of siderophile elements. However, the concentrations of highly siderophile elements, as well as the moderately volatile elements Te and Se in the silicate Earth are higher than expected based on partition coefficients and it was suggested that a large portion of these elements was added after core formation by a late veneer (e.g., Kimura et al., 1974; Rose-Weston et al., 2009; Wang and

Becker, 2013). Based on Se/Te ratios, Wang and Becker (2013) concluded that the late veneer had a composition similar to CI or CM carbonaceous chondrites. However, near-chondritic Se/Te ratios of Earth's mantle may not be a primary signature, as was suggested by König et al. (2014).

The new Te stable isotope data presented here demonstrate that different chondrite groups display differences in their $\delta^{130/125}$Te isotope composition. In order to use Te stable isotope fractionation data to deduce information regarding the building blocks of the Earth and the composition of the late veneer, one would need to also know the Te isotope composition of the silicate Earth. The silicate Earth has two orders of magnitude lower Te contents compared to meteorites (e.g., McDonough and Sun, 1995) and hence such analyses are analytically challenging and would require further improvement of the methodologies described here. Further studies will be necessary to investigate the Te stable isotope composition of the silicate Earth. However, assuming that only small stable isotope fractionation is produced on Earth the available Te stable isotope fractionation data for terrestrial samples and standards may provide an estimate for the Te composition of the silicate Earth. The terrestrial samples and standards have a Te stable isotope composition ranging from -0.15 ± 0.07 to +0.74 ± 0.05 for $\delta^{130/125}$Te (Table 1). In detail the shales may provide a useful guide to the composition of the silicate Earth as they potentially represent an average crustal composition derived by weathering and they display a slightly narrower range in $\delta^{130/125}$Te with -0.08 ± 0.08 and 0.42 ± 0.01, which overlaps with compositions determined for the CM and CI carbonaceous chondrites (Table 1, 2, Fig. 3b). By contrast, significantly more negative Te isotope compositions of -0.55 ± 0.07 to -0.24 ± 0.08 were determined for the studied CV and enstatite chondrites. The CO carbonaceous chondrite Lance ($\delta^{130/125}$Te = -0.35 ± 0.20) also displays a more negative Te isotope signature compared to the terrestrial samples, whereas the Te isotope data for the CO chondrite Ornans ($\delta^{130/125}$Te = -0.18 ± 0.26) overlaps with the compositional range determined for terrestrial samples. Ordinary chondrites

display a large range in Te stable isotope fractionation, although their average composition overlaps though with that of terrestrial samples ($\delta^{130/125}$Te = -0.4 ± 4.1). Therefore, the presented data are in agreement with Te derived from a late veneer that has a composition similar to that of CI or CM carbonaceous chondrites, a conclusion also reached by Wang and Becker (2013) based on Se/Te and chalcogen/Ir ratios. However, the recent Ru nucleosynthetic isotope study by Fischer-Gödde and Kleine (2017) indicates that the late veneer has a non-chondritic composition that is most similar to enstatite chondrites. Further work will be necessary to improve our understanding of the behavior of Te during core formation to constrain the late veneer contribution to the Te budget of the silicate Earth and to characterize the Te stable isotope signature of the silicate Earth to deduce whether the Ru and Te isotope data provide contradicting evidence to the composition of the late veneer.

6. CONCLUSIONS

Significant Te stable fractionation is present in terrestrial sediments of 0.85‰ for $\delta^{130/125}$Te indicating that Te isotopes have a potential to become a sedimentary geochemical proxy. Geochemical exploration reference samples display 0.6 ‰ variations in $\delta^{130/125}$Te.

Tellurium displays large isotope fractionation in unequilibrated ordinary chondrites with an overall variation of 6.3‰ for $\delta^{130/125}$Te, similar to some other moderately and highly volatile elements (Luck et al., 2005; Nebel et al., 2011; Schönbächler et al., 2008; Wombacher et al., 2008; Wombacher et al., 2003). The observed Te isotope fractionations in ordinary chondrites is likely caused by evaporation and condensation processes during parent-body metamorphism or alternatively potentially in the nebula or associated with chondrule formation.

Smaller compositional variations in the Te stable isotope composition are present between different groups of carbonaceous chondrites and between enstatite and carbonaceous

chondrites, indicating the mixing of two or more components with distinct Te isotope compositions reflecting likely Te isotope fractionation in the early solar system. Parent body processes could also have affected the Te stable isotope signature, as is indicated by the sample heterogeneity of Orgueil. Tellurium isotope variations within carbonaceous and enstatite chondrites display a correlation with the oxidation state of samples and hence might provide a nebular record of the environment where the different groups of chondrites formed.

The Te stable isotope composition of terrestrial samples is more positive in $\delta^{130/125}$Te compared to enstatite and CV carbonaceous chondrites. If the silicate Earth has a similar Te isotope signature to the terrestrial samples and standards investigated in this study, then the present data would be in agreement with a late veneer contribution with composition similar to that of CI, CM or potentially CO chondrites. Future work to determine the Te stable isotope signature of the silicate Earth is necessary though to verify this conclusion.

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FIGURE CAPTIONS

Fig. 1. Propagation of uncertainties of $\delta^{130/125}$Te in relation of the spike proportion in the spike-sample mixture.

Fig 2. $\delta^{130/125}$Te and Te concentration data for terrestrial samples. Closed symbols: sediment samples, open symbols: USGS geochemical exploration reference samples. Uncertainties for Te concentrations are smaller than symbols. For $\delta^{130/125}$Te displayed errors are 2 SD of repeat analyses (see Tables 1, 2). For single analyses the uncertainty is the daily reproducibility (2 SD) of the standard.

Fig. 3. $\delta^{130/125}$Te data for chondritic meteorites, terrestrial samples and standard solutions (see Tables 1, 2). Panel a) all chondrite data, b) all data beside Mezö-Madaras (L3.7) and Dhajala (H3.8). Uncertainties for individual analyses are the daily reproducibility (2 SD) of the standard. For standard solutions, uncertainties are 2 SD of repeat analyses. Different symbols reflect different sample digestions. The grey area marks the composition of the terrestrial data.

Fig. 4. Tellurium stable isotope fractionation data and Te concentrations for chondrites. Panel a) ordinary chondrite data, b) all data beside Mezö-Madaras (L3.7) and Dhajala (H3.8). Uncertainties for Te concentrations are smaller than symbols, errors for $\delta^{130/125}$Te are 2 SD of repeat analyses (Table 1). For single analyses uncertainties are the daily reproducibility (2 SD) of the standard.

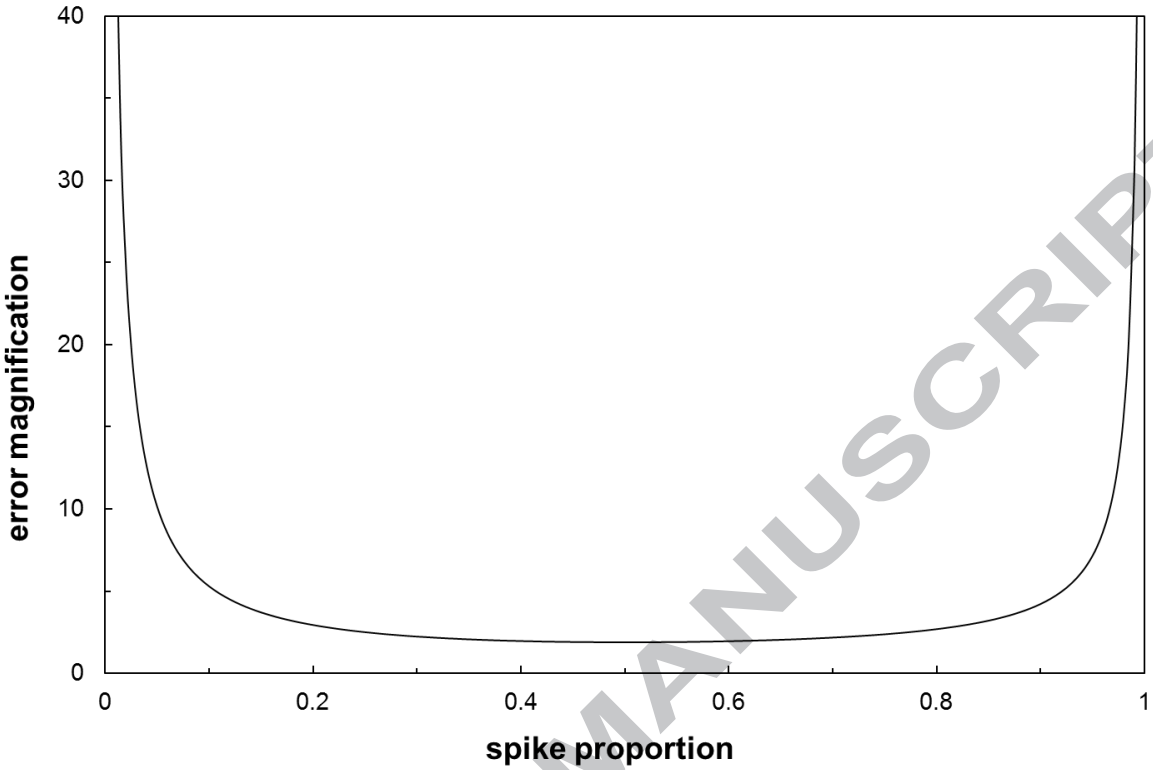

Fig. 1

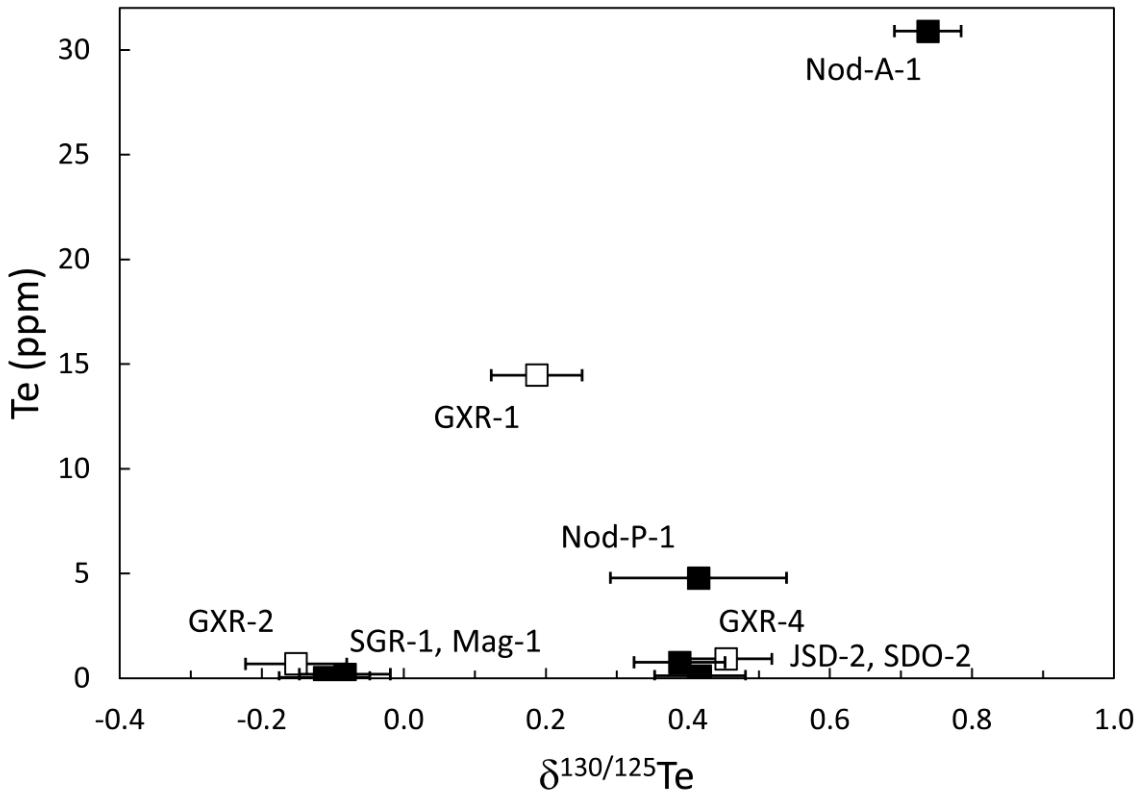

Fig. 2

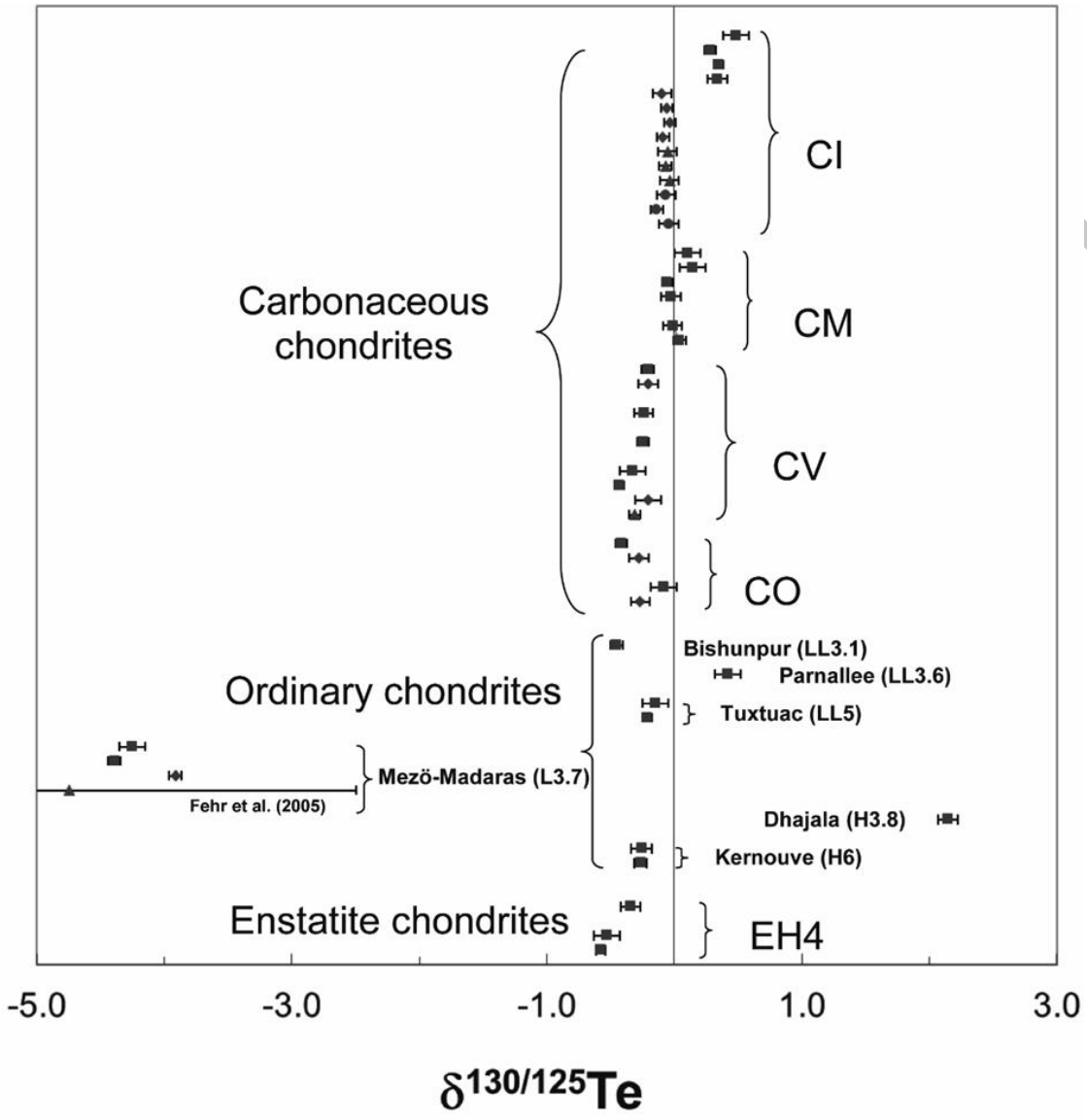
Carbonaceous chondrites
CI
CM
CV
CO
Ordinary chondrites
Bishunpur (LL3.1)
Parnallee (LL3.6)
Tuxtuac (LL5)
Mezö-Madaras (L3.7)
Fehr et al. (2005)
Dhajala (H3.8)
Kernouve (H6)
Enstatite chondrites
EH4
-5.0
-3.0
-1.0
1.0
3.0
δ130/125Te

Fig. 3a

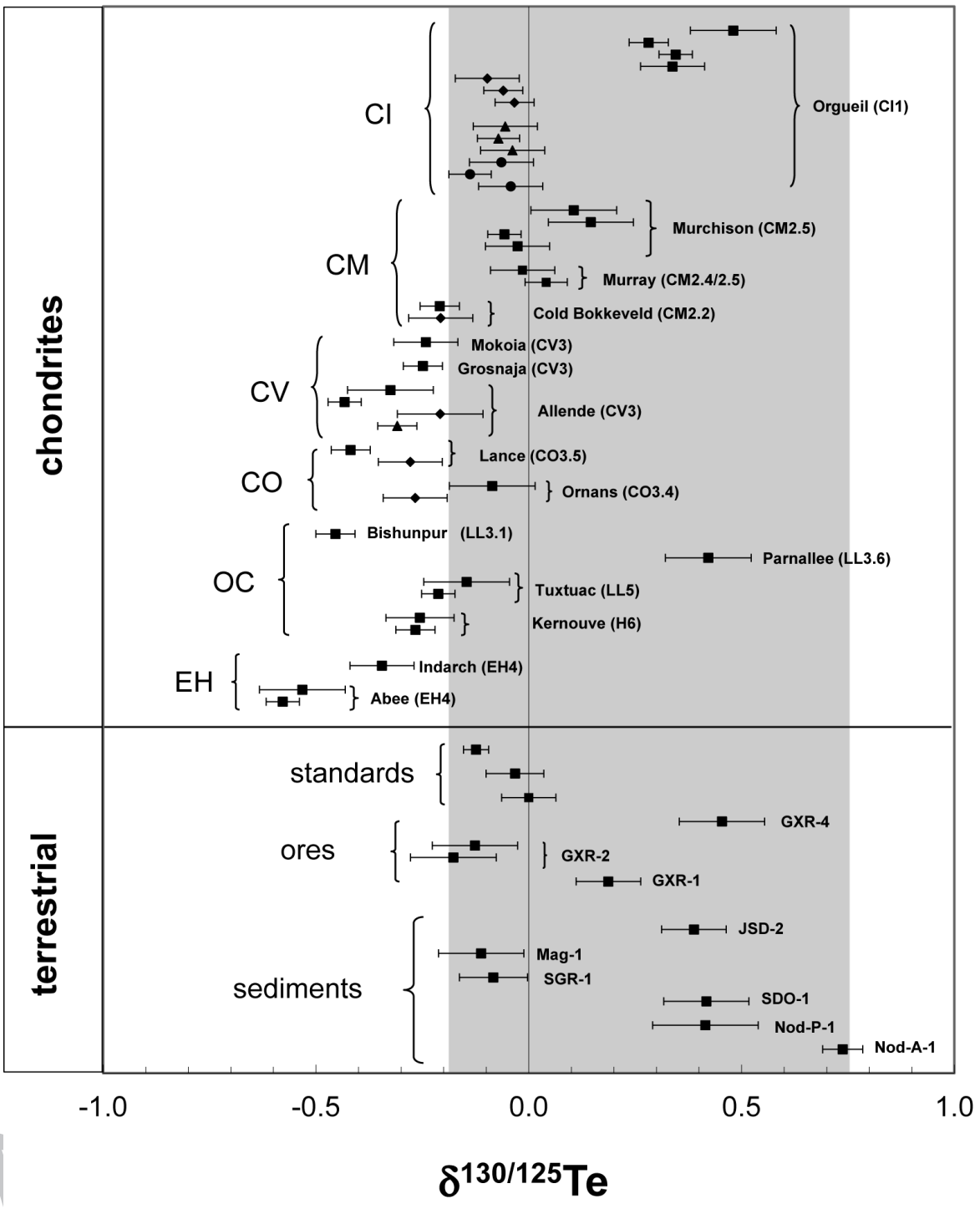
chondrites
terrestrial
CI
Orgueil (CI1)
CM
Murchison (CM2.5)
Murray (CM2.4/2.5)
Cold Bokkeveld (CM2.2)
CV
Mokoia (CV3)
Grosnaja (CV3)
Allende (CV3)
CO
Lance (CO3.5)
Ornans (CO3.4)
OC
Bishunpur (LL3.1)
Parnallee (LL3.6)
Tuxtuac (LL5)
Kernouve (H6)
EH
Indarch (EH4)
Abee (EH4)
standards
ores
GXR-4
GXR-2
GXR-1
sediments
JSD-2
Mag-1
SGR-1
SDO-1
Nod-P-1
Nod-A-1
-1.0
-0.5
0.0
0.5
1.0
δ130/125Te

Fig. 3b

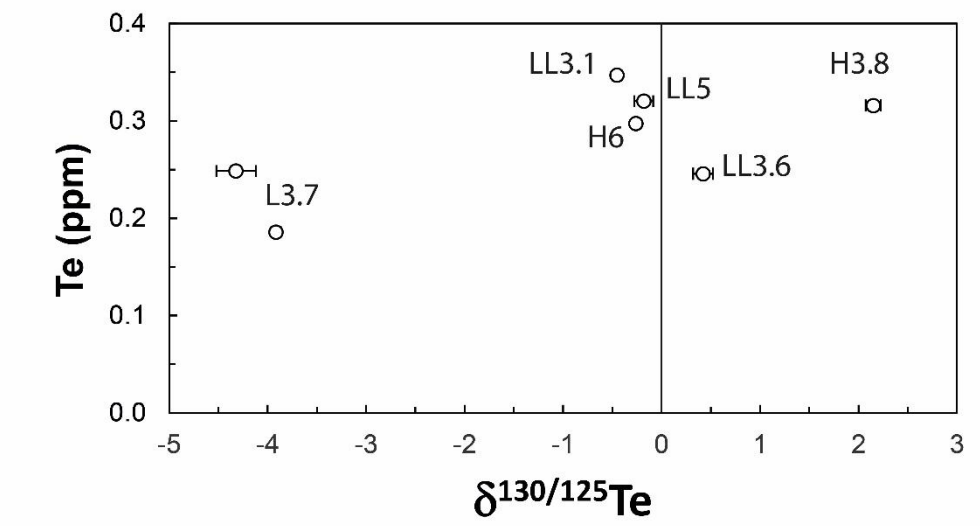

Fig. 4a

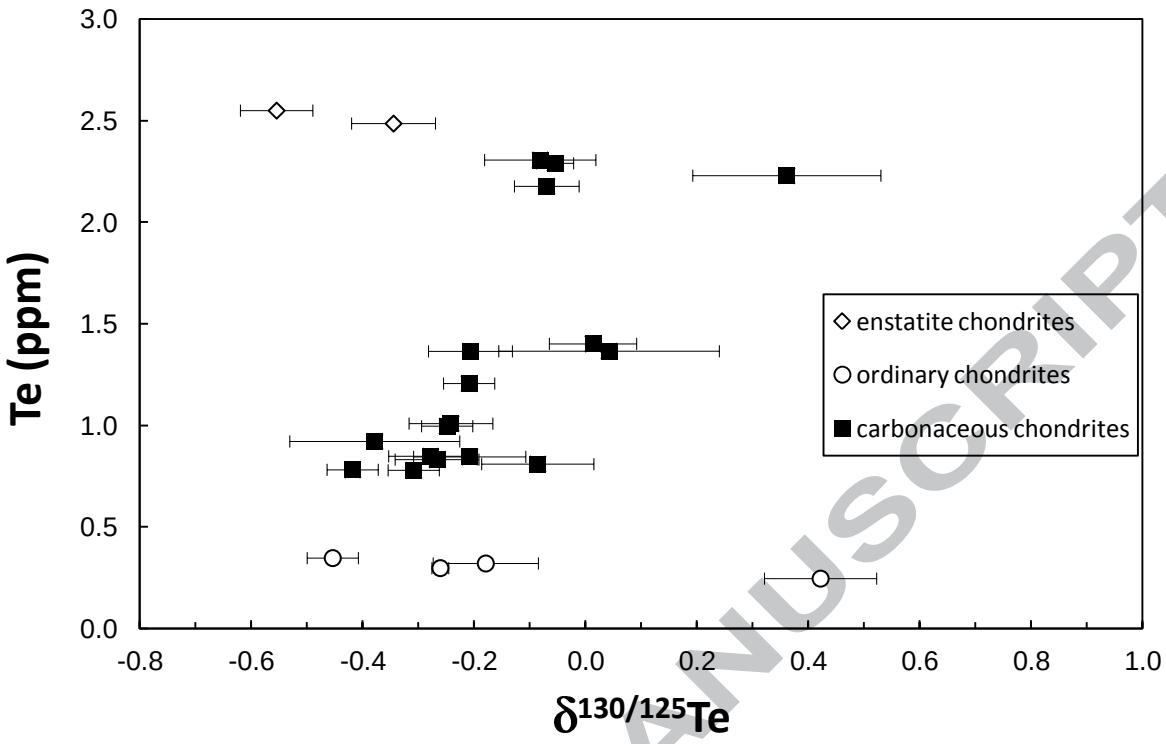

Fig. 4b

Table 1: $\delta^{130/125}$Te Te stable isotope data of terrestrial samples and standards and Te concentration data

Sample name	Supplier	Description		#[a]	$\delta^{130/125}$Te[b]	2SD[c]	Te (ppm)[d]	Te (ppm) literature
Nod-A-1			rep. 1		0.72			
			rep. 2		0.73			
			rep. 3[i]		0.76			
Mean Nod-A-1	USGS	Manganese nodule, Atlantic ocean		3	0.74	0.05	n.d.	30.9[e]
Nod-P-1			rep. 1[i]		0.36			
			rep. 2[i]		0.33			
			rep. 3		0.39			
			rep. 4		0.48			
			rep. 5		0.47			
			rep. 6		0.45			
Mean Nod-P-1	USGS	Manganese nodule, Pacific ocean		6	0.42	0.12	n.d.	4.8[e]
SDO-1	USGS	devonian Ohio shale			0.42	0.10	0.12	
SGR-1	USGS	green river shale			-0.08	0.08	0.20	0.23[f]
Mag-1	USGS	marine mud, Gulf of Maine, Atlantic			-0.11	0.10	0.05	0.07[g]
JSD-2	GSJ	stream sediment			0.39	0.08	0.77	0.8[h]
GXR-1	USGS	jasperoid, Drum mountains, Utah			0.19	0.08	14.5	16[h]
GXR-2			rep. 1		-0.18		0.69	
			rep. 2		-0.13		0.69	
Mean GXR -2	USGS	soil, Park City, Utah		2	-0.15	0.07	0.69	0.75[h]
GXR-4	USGS	porphyry copper mill heads, Utah			0.45	0.10	0.93	1.06[h]
Standards solutions								
Mean Te Alfa Aesar metal standard				154	0.00	0.06		
Mean Te standard SRM 3156				9	-0.12	0.03		
Te Alfa Aesar solution standard				2	-0.03	0.07		
Column processed standard								
300 ng Te -1			rep. 1		0.09			
			rep. 2		0.09			
			rep. 3		0.09			
			rep. 4		0.09			
Mean 300 ng Te -1		Te Alfa Aesar metal		4	0.09	0.00		
300 ng -2		Te Alfa Aesar metal			0.11	0.08		
50 ng Te		Te Alfa Aesar metal			0.02	0.08		
Mean Te		Te Alfa Aesar metal		3	0.07	0.10		

Te double spike was added after sample digestion

[a] number of individual analyses: total number of repeat measurements (rep.)

[b] $\delta^{130}Te/^{125}Te = [(^{130}Te/^{125}Te_{sample}/ {}^{130}Te/^{125}Te \text{ alfa aesar metal})-1] \times 1000$

[c] 2SD reproducibility of daily standards shown for single analyses

[d] 2SE of single analyses and 2SD of repeat analyses are <0.001 ppm and are not displayed.

[e] Axelsson et al. (2002) [f] Hall [g] Wang et al. (2015) [h] Terashima (2001)

[i] Analysis performed using standard skimmer cone

Table 2: $\delta^{130/125}$Te Te stable isotope fractionation and Te concentration data of chondrites

Sample name	Classification	Specimen	Shock		#[a]	$\delta^{130/125}$Te[b]	2SD[c]	Te (ppm)	2SD[d]	Te (ppm) literature
Orgueil	CI			diss. 1, rep. 1		0.48		2.23		
				diss. 1, rep. 2		0.28		2.23		
				diss. 1, rep. 3		0.35		2.23		
				diss. 1, rep. 4		0.34		2.23		
				Mean diss. 1[e]	4	0.36	0.17	2.23		
				diss. 2, rep. 1		-0.10		2.18		
				diss. 2, rep. 2		-0.06		2.18		
				diss. 2, rep. 3		-0.03		2.18		
				diss. 2, rep. 4		-0.09		2.18		
				Mean diss. 2	4	-0.07	0.06	2.18		
				diss. 3, rep. 1		-0.06		2.29		
				diss. 3, rep. 2		-0.07		2.29		
				diss. 3, rep. 3		-0.04		2.29		
				Mean diss. 3[e]	3	-0.05	0.03	2.29		
				diss. 4, rep. 1		-0.06		2.31		
				diss. 4, rep. 2		-0.14		2.31		
				diss. 4, rep. 3		-0.04		2.31		
				Mean diss. 4[e]	3	-0.08	0.10	2.31		
Mean, Orgueil	CI				4	0.04	0.42	2.25	0.12	2.3 - 2.6[f-i]
Cold Bokkeveld	CM2.2	BM82818		diss. 1		-0.21	0.05	1.21		
		BM82818		diss. 2[e]		-0.21	0.08	1.37		
Mean Cold Bokkeveld	CM2.2		S1		2	-0.21	0.00	1.29	0.22	1.1[j]- 1.3[k]
Murray	CM 2.4/2.5	BM1971, 288		rep. 1		-0.01		1.40		
				rep. 2		0.04		1.40		
Mean Murray	CM 2.4/2.5		S1	[e]		0.01	0.08	1.40		1.4[f]
Murchison	CM2.5	1988.M23		rep. 1		0.11		1.37		
				rep. 2		0.15		1.37		
				rep. 3		-0.06		1.37		
				rep. 4		-0.03		1.36		
Mean Murchison	CM2.5		S1-2	[e]	4	0.04	0.20	1.37		1.3-1.8[f-i, l]
Ornans	CO3.4	BM1920, 329		diss. 1		-0.09	0.10	0.81		
		BM1920, 329		diss. 2[e]		-0.27	0.08	0.83		
Mean Ornans	CO3.4		S1		2	-0.18	0.26	0.82	0.03	1[m]
Lance	CO3.5	BM1985, M153		diss. 1		-0.42	0.05	0.78		
		BM1985, M153		diss. 2[e]		-0.28	0.08	0.85		
Mean Lance	CO3.5		S1		2	-0.35	0.20	0.81	0.09	0.84[n]
Allende	CV3	BM1969, 148		diss. 1, rep. 1		-0.32		0.92		
				diss. 1, rep. 2		-0.43		0.92		
				Mean diss. 1[e]	2	-0.38	0.15	0.92		
		BM1969, 148		diss. 2		-0.21	0.10	0.85		
		BM1969, 148		diss. 3		-0.31	0.05	0.78		
Mean Allende	CV3		S1		3	-0.30	0.17	0.85	0.14	0.86 - 1.1[f,h,i,l,o]

Table 2 continued

Sample name	Classification	Specimen	Shock		#[a]	$\delta^{130/125}$Te[b]	2SD[c]	Te (ppm)	2SD[d]	Te (ppm) literature
Grosnaja	CV3	63624	S3			-0.25	0.05	1.00		1.2[n]
Mokoia	CV3	BM 1910.729	S1	[e]		-0.24	0.08	1.01		1[n]
Parnallee	LL3.6	BM34792	b			0.42	0.10	0.25		0.2[p]
Bishunpur	LL3.1	BM80339	S2			-0.45	0.05	0.35		0.5[p]
Tuxtuac	LL5	BM1981, M7		rep. 1		-0.15		0.32		
				rep. 2		-0.21		0.32		
Mean Tuxtuac	LL5	BM1981, M7	S2	[e]	2	-0.18	0.09	0.32		
Mezö-Madaras	L3.7	BM90270		diss. 1, rep. 1		-4.25		0.25		
				diss. 1, rep. 2		-4.39		0.25		
				Mean diss. 1[e]	2	-4.32	0.20	0.25		
		BM90270		diss. 2		-3.91	0.05	0.19		
Mean Mezö-Madaras	L3.7		b		2	-4.12	0.58	0.22	0.09	0.57[j]- 0.88[q]
Dhajala	H3.8	BM1976, M12	S1	[e]		2.15	0.08	0.32		
Kernouve	H6	BM43400		rep. 1		-0.26		0.30		
				rep. 2		-0.27		0.30		
Mean Kernouve	H6		S1		2	-0.26	0.02	0.30		0.34[r]-0.38[s]
Abee	EH4	BM1992, M7		rep. 1		-0.53		2.55		
				rep. 2		-0.58		2.55		
Mean Abee	EH4		S2-4	[e]	2	-0.55	0.07	2.55		1.7[f] - 3[i]
Indarch	EH4	BM86948	S3	[e]		-0.34	0.08	2.49		2[t]

[a] number of individual analyses: total number of repeat measurements (rep.) of one or several separate sample dissolutions (diss.)
[b] $\delta^{130}Te/^{125}Te = [(^{130}Te/^{125}Te_{sample}/ ^{130}Te/^{125}Te \text{ alfa aesar metal})-1] \times 1000$
[c] 2SD reproducibility of daily standards shown for single analyses
[d] 2SD of repeat analyses are <0.002 ppm and are not displayed
[e] Te double spike was added before sample digestion
Literature data are from [f] Smith et al. (1977), [g] Xiao and Lipschutz (1992), [h] Friedrich et al. (2002), [i] Fehr et al. (2005), [j] Wolf et al. (1980), [k] Zolensky et al. (1997), [l] Makishima and Nakamura (2009), [m] Takahashi et al. (1978), [n] Anders et al. (1976), [o] Wang et al. (2015), [p] Binz et al. (1976), [q] Keays et al. (1971), [r] Morgan et al. (1985), [s] Lingner et al. (1978), [t] Kallemeyn and Wasson (1986)